PATENT SPECIFICATION

NO DRAWINGS

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COMPLETE SPECIFICATION

Magnesium Antimonate Phosphor

We, SYLVANIA ELECTRIC PRODUCTS INC., a corporation organised under the laws of the State of Delaware, United States of America, of 100 West 10th Street, Wilmington, Delaware, United States of America, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

This invention relates to magnesium lithium antimonate phosphors.

Such phosphors activated by either manganese or uranium, or both, are known from the specification of Patent No. 707,101, but their fluorescent emission is in the green to red region of the spectrum.

It has been discovered that the emission can be shifted to the blue by replacing the manganese and/or uranium activators by tungsten or titanium.

In addition, it has been found that the use of titanium as a secondary activator, with manganese or uranium as the other activator, will greatly increase the light emission over that obtained with activation by manganese or uranium alone.

The proportion by weight of magnesium, lithium and antimony, expressed as the oxides, in the phosphor, can be between 20% and 60% of magnesium oxide (MgO), between 5% and 35% of lithium oxide (Li_2O), and between 30% and 70% of antimony trioxide (Sb_2O_3).

It is preferred, however, to use between 25% and 40% magnesium oxide, between 10% and 20% lithium oxide and between 45% and 60% antimony trioxide.

In the preparation of these phosphors, it is preferred to use basic magnesium carbonate, lithium carbonate and antimony trioxide as the raw materials. However, substitutions of other raw materials will give substantially the same results, these substitutions being obvious to those skilled in the art. Thus, for example, magnesium oxide or magnesium hydroxide may be used in place of the basic magnesium carbonate and similarly lithium oxide or lithium hydroxide may be used in place of the lithium carbonate.

The concentration of titanium or tungsten activators may vary over a considerable range from 0.0001 to 0.25 gram atoms of activator per 100 grams of combined oxides in the original mixture. Preferably however the best phosphors are obtained with titanium concentrations between 0.05 and 0.04 gram atoms per 100 grams of combined oxides and with tungsten concentrations between 0.001 and 0.02 gram atoms per 100 grams of combined oxides. It is preferred, for convenience, to use titanium dioxide or hydrated tungsten trioxide, but halides or sulphates may also be employed. The effect of titanium activator concentration for a specific matrix composition is shown by way of example in Table I.

[*Price 3s. 6d.*]

TABLE I

Effect of Titanium Concentration

Matrix Composition: MgO, 35%; Li_2O, 15% and Sb_2O_3, 50% by weight

Gram Moles of TiO_2 per 100 g. combined oxides	Relative Brightness for Final Firing Temp of 2050° F.	2200° F.
0.0025	12	29
0.0075	23	29
0.020	38	46
0.025	45	53
0.035	43	38
0.050	42	34
0.075	41	18

The effect of tungsten activator concentration for a specific matrix composition is shown by way of example in Table II.

TABLE II

Effect of Tungsten Concentration

Matrix composition: MgO, 35%; Li_2O, 15% and Sb_2O_3, 50% by weight.

Gram Moles of WO_3H_2O per 100 g. combined oxides	Relative Brightness for Final Firing Temp. of 2050° F.	2200° F.
0.0002	16	22
0.002	33	21
0.004	33	38
0.010	30	25
0.016	31	23
0.032	21	21

The optimum firing temperature depends upon the activator concentration as shown in Tables I and II and to some extent on the composition of the matrix. At any given temperature compositions which are high in antimony trioxide or lithium oxide give harder products and, since hard products are undesirable in coating lamps, such compositions require lower firing temperatures, thus giving softer products. However, it has been found that the range from 1800° to 2200° F. is satisfactory for most compositions.

While a single firing of the well mixed raw materials gives a useful phosphor, the output is generally increased by dry grinding the product, mixing well and firing a second time. It is not necessary to employ the same temperature for both firings and, in fact, it is

preferred to use a temperature of about 1800° to 2000° F. for the first firing and then to refire at a higher temperature.

If the magnesium lithium antimonate phosphor is doubly activated by titanium and manganese the red output of the phosphor is increased relative to the manganese-only-activated phosphor as shown by the data of Table III.

TABLE III

Red Response of Mn and (Mn + Ti) — Activated Altimonates

Firing Procedure	Relative Red Response to 3650 A		% Increase of Doubly Activated Phosphor
	Mn Activated	(Mn+Ti) Activated	
1 hour at 1925° F.	18	19	6
1 hour at 2100° F.	27	36	33
1 hour at 2200° F.	29	42	45

The Mn is present to the extent of about 0.1% by weight of the phosphor in the above table, and the titanium is present to the extent of about 2% on the same basis. The firing procedure is as previously described.

Titanium will also act as a secondary activator in the uranium-activated magnesium lithium antimonate phosphor resulting in a net increase in the emission in the green. For example a phosphor prepared with 1.0 gram of uranyl nitrate and 1.0 gram of titanium dioxide with 50 grams of combined oxides is approximately 19% brighter in the green than the same phosphor without TiO_2.

The uranium is present to the extent of about 2% by weight of the phosphor and the titanium to the extent of about 2% on the same basis. The firing procedure is as previously described.

Specific examples will now be given of the preparation of phosphors according to the invention.

EXAMPLE 1.

A phosphor was prepared for which the matrix had the composition: MgO, 0.563 gram moles; Li_2O, 0.326 gram moles, Sb_2O_3, 0.111 gram moles and in which the titanium activator concentration was 0.0162 gram atoms per 1.0 gram moles of combined oxides. Raw materials as listed below were carefully weighed, using the highest purity materials commercially available.

Basic magnesium carbonate	80.7 grams	(assay 43.3% MgO)
Lithium carbonate	37.0	(assay 40.5% Li_2O)
Antimony trioxide	50.0	(assay 100% Sb_2O_3)
Titanium dioxide	2.0	(assay 100% TiO_2)

These materials were dry mixed by tumbling and then transferred to fused silica crucibles and fired for 1 hour at 1950° F. The product was crushed after cooling, well mixed and after transfer to a silica crucible fired again for 1 hour at 2150° F.

EXAMPLE 2.

The W-activated phosphor was prepared with the same matrix composition but with a tungsten activator concentration of 0.0026 gram atoms per 1.0 gram moles of combined oxides. Thus the raw materials as listed below were carefully weighed, using the highest purity materials commercially available.

Basic magnesium carbonate	80.7 grams (assay 43.3% MgO)
Lithium carbonate	37.0 grams (assay 40.5% Li_2O)
Antimony trioxide	50.0 grams (assay 100% Sb_2O_3)
Tungstic acid	1.0 gram (assay 91.9% WO_3)

These materials were dry mixed by tumbling and then transferred to fused silica crucibles and fired for 1 hour at 1950° F. The product was crushed after cooling, well mixed and after transfer to a silica crucible fired again for 1 hour at 2150° F.

WHAT WE CLAIM IS:—

1. A phosphor comprising magnesium lithium antimonate activated by tungsten or titanium.

2. A phosphor according to claim 1, containing as further activator manganese and/or uranium.

3. A phosphor according to claim 1 or 2, wherein the quantity of tungsten or titanium is between 0.0001 and 0.25 gram atoms per 100 grams of phosphor.

4. A phosphor substantially as hereinbefore described.

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